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Diagnostic development for the ElectriCOIL flow system

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ABSTRACT

Detailed studies of mechanisms for producing electrically initiated COIL lasers were previously presented. Results of those studies along with more recent experimental results show that electric excitation is a very complex process that must be investigated with advanced diagnostics. Theoretical studies indicate that fractions of $O_2(^1\Delta)$ may be produced in the discharge that will permit lasing of an ElectriCOIL system. Recent kinetic studies indicate a range of useful operating parameters for ElectriCOIL that are analogous to those achieved in the all-chemical device. This can be accomplished at E/N's in the range of 10^{-16} Volt-cm^2. An experimental test bed has been built up to allow detailed diagnostic measurements of the discharge efficiencies and other experimental parameters. Results of early experiments are presented.

Keywords: chemical oxygen-iodine laser, COIL, ElectriCOIL, RF excitation of oxygen, singlet-delta oxygen

1. INTRODUCTION

Researchers at CU Aerospace (CUA) and University of Illinois at Urbana-Champaign (UIUC) have been operating clemical lasers (HF/DF, Overtone HF, and COIL) for a number of years. The classical chemical oxygen-iodine laser (COIL) [McDermott, 1978] operates on the electronic transition of the iodine atom at 1315 nm, $I^*(^2P_{1/2}) \rightarrow I(^2P_{3/2}) + h\nu$. The population inversion is maintained by the near resonant energy transfer between the excited singlet oxygen $O_2(^1\Delta)$ molecule and the I atom ground state $I(^2P_{3/2})$ as follows: $O_2(^1\Delta) + I(^2P_{3/2}) \rightarrow O_2(^3\Sigma) + I(^2P_{1/2})$. Traditionally, this pumping reaction has been fed by a liquid chemistry singlet oxygen generator (SOG). Workers at CUA and UIUC are now addressing the engineering issues associated with an electrical COIL system (**ElectriCOIL**) [Carroll, 2001; King, 2001].

AFRL demonstrated that significant quantities of excited iodine atoms can be produced using an all gas phase generator. The excited species which transfers its energy to Iodine atoms, in this case $NCl(^1\Delta)$, can be produced without a liquid phase and has provided a recent lasing demonstration (AGIL) [Henshaw, 2000]. We believe that it is possible to construct a highly efficient electric generation scheme to provide the precursor energy donor species $O_2(^1\Delta)$ and that the ElectriCOIL concept can subsequently be realized.

Workers in Japan [Itami, 1999], Russia [Ivanov, 1999], and in the U.S. [King, 2001] have shown that flowing discharge tubes containing ground state oxygen can produce significant quantities of the desired, $O_2(^1\Delta)$ precursor molecules. We believe this work suggests that one can transform such research into a practical laser system. Atomic iodine injection rather than molecular iodine injection will give added benefit to the ElectriCOIL laser. Figure 1 illustrates the limitations of existing COIL technology as well as the possible improvement from the implementation of ElectriCOIL technology. An electronically produced $O_2(^1\Delta)$ generator could be even more effective if implemented simultaneously with atomic iodine injection.

Fujii [Fujii, 1994] reported good success, 17% yield of $O_2(^1\Delta)$, with a small RF generator. More recently, workers in Japan [Itami, 1999] from Fujisaki Electric provided some evidence that they could produce 21% $O_2(^1\Delta)$ in a microwave discharge. The experiments were carried out in subsonic axial flow at 2 Torr in a 30 cm long tube with window attachments for observation. Hill [Hill, 2001] reported a value of 16%. Schmiedberger [Schmiedberger, 2001] reported a 32% yield under low-pressure conditions (0.43 Torr) with an RF discharge. We have recently obtained an $O_2(^1\Delta)$ yield of ≈16% in our flowing RF discharge experiments at a pressure of 2 Torr and ≈15% of 3.6 Torr.

We choose chemical efficiency as a way of relating classic COIL performance to our ElectriCOIL concept. Even an ElectriCOIL that provides 20% chemical efficiency (shown in Fig. 1) is an enormous improvement to the classic COIL

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design because it will lead to a significant reduction of weight on a large weapons class laser, simpler operation procedures, and reduce the overall risk.

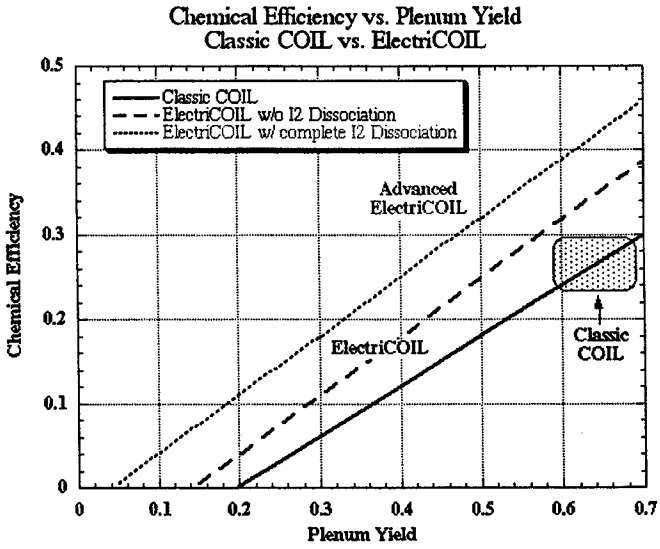

Fig. 1. A comparison between classic COIL technology and the performance possibilities for an ElectriCOIL device are illustrated here. Results based upon a heuristic equation [Hon, 1996].

Calculations using the Blaze II chemical laser model [Sentman, 1977; Carroll, 1995] support the qualitative conclusions drawn from a heuristic equation [Hon, 1996], Figs. 1 and 2. The advantage of the ElectriCOIL concept is immediately realized when examining the gain curve with and without molecular iodine pre-dissociation, Fig. 3. The gain curve, with a 20% yield and no pre-dissociation, is very low and would not make an efficient laser. However, when the molecular iodine is completely pre-dissociated, then the gain curve, even with a 20% yield, compares very favorably with the classic COIL type gain curve having a 67% yield.

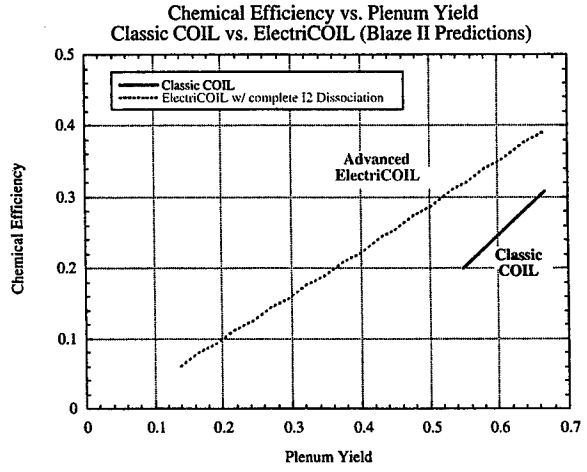

Figure 2. Blaze predictions comparing classic COIL technology and the performance possibilities for an ElectriCOIL device are illustrated. While ElectriCOIL may not be able to match the plenum yield of today's COIL system, significantly smaller yields are capable of matching current chemical efficiencies.

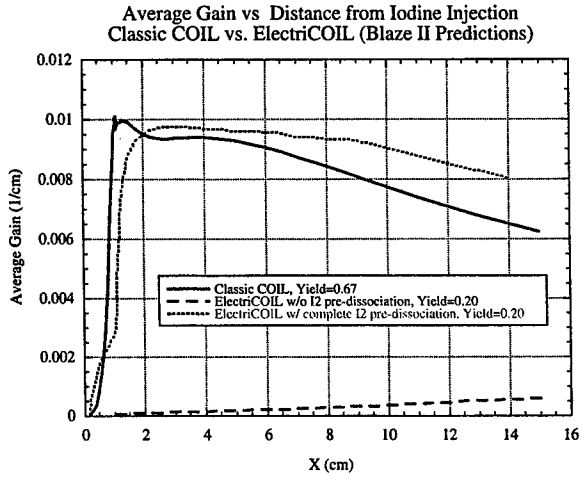

Fig. 3. Blaze predictions of the average gain as a function of distance from the iodine injection position for classic COIL with a yield of 0.67, ElectriCOIL with and without molecular iodine pre-dissociation. Helium diluent was used for these calculations.

Figure 3 also illustrates the innovative concept of combining both electrically produced singlet delta oxygen and pre-dissociated iodine. Each of these concepts have been demonstrated individually, but not in combination; when both ideas are implemented together it is possible to obtain significant gain and laser power with a mere 20% yield; this yield has already been generated in experiments conducted by Schmiedberger [Schmiedberger, 2001]. Experimental work in the area of iodine pre-dissociation has been conducted by Endo and Fujioka's group in Japan [Endo, 1999]. They reported nearly total dissociation from interaction of an Iodine/N_2 stream within the microwave cavity. Iodine pre-dissociation has also been investigated using three-dimensional CFD computations by Madden *et al.* [Madden, 1998]; Madden's results indicated that the injection of atomic iodine slightly downstream of the throat would enhance the power output of a classic COIL device. Recently, CUA and UIUC implemented an LIF experiment that showed 50% dissociation downstream from a dc electric discharge and about 95% in an RF discharge.

2. THEORETICAL BACKGROUND AND MODELING

The motivation for the use of an electrical discharge for the production of $O_2(^1\Delta)$ and $O_2(^1\Sigma)$ states is provided by Figures 4 and 5. These are the predictions from a Boltzmann Equation solver [Kushner, 2000] which tracks the fraction of electrical power utilized for each electron energy loss process. Shown here is the fraction used to excite the $O_2(^1\Delta)$ state (0.977 eV/exc.) and the fraction used to excite the $O_2(^1\Sigma)$ state (1.627 eV). In both the pure O_2 and 1:1 mixture of Helium and O_2, nearly 50% of the electrical power can be used to produce $O_2(^1\Delta g)$ molecules and 20% can be used for the production of the $O_2(^1\Sigma g)$ state at an E/N of about $8x10^{-17}$ volt-cm^2.

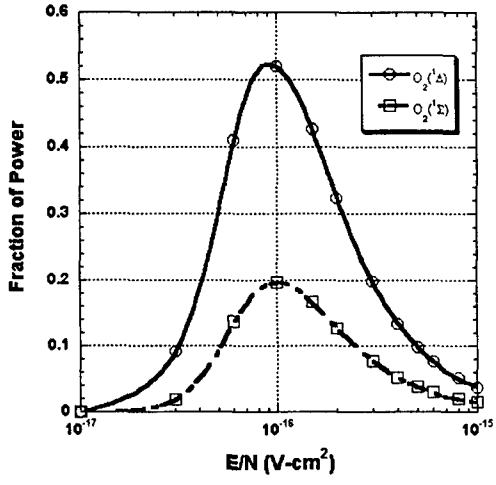

Figure 4: Results of Bolztmann calculation for pure O_2.

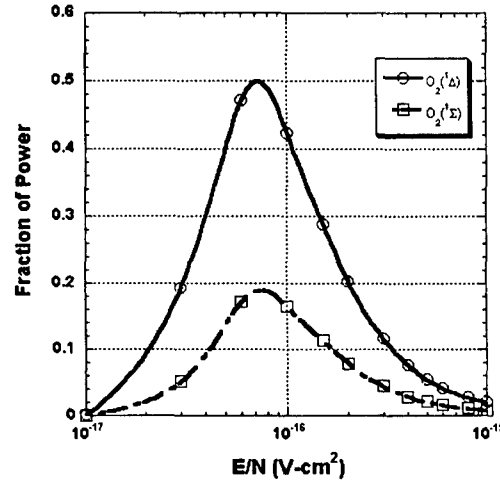

Figure 5: Boltzmann calculation with mix of He:O_2 = 1:1.

Obviously, the efficiency and yield of $O_2(^1\Delta_g)$ molecules are critically dependent on the field (E) to neutral gas density N, or E/N, with the dependence of the use of electrical power shown in Figures 4 and 5. Although these graphs show a clear maximum at E/N ~ 10^{-16} V-cm^2, there is no guarantee that the discharge will, in fact, operate at that value. Fortunately, experiments with a simple DC discharge (upstream from the RF one) indicated an E/N of ~1.5 - $2.0x10^{-16}$ V-cm^2 and obeyed the simple scaling laws for the positive column. Thus we feel that this is a reasonable estimate for spatially averaged value for the induction discharge used in our latest experiments. In such a discharge, the field is a maximum at the walls and zero on the axis. Recent calculations suggest an effective E/N of $1.2x10^{-16}$ V-cm^2 at 4.2 Torr and 300 Watts.

Thus, if we assume that 45% of the baseline 300 Watts of RF power is used for the production of $O_2(^1\Delta_g)$ in the cylindrical tube (R=2.54 cm and length of 10 cm) with a volume of 200 cm^3 at 4.2 torr of pure O_2, then the production rate for $O_2(^1\Delta_g)$ is:

$$\frac{d[^1\Delta_g]}{dt} = +\frac{135W}{Vol. = 200cm^3} \times \frac{1}{0.9775eV} \times \frac{1}{1.6\times10^{-19} J/eV} = 4.27x10^{18} cm^{-3}/s. \quad (1)$$

For the conditions used in Fig. 10, the flow velocity is approximately $2x10^3$cm/s and thus the lifetime of the molecules in the discharge region is 5 ms. Thus, this simple theory would predict an exit density of $O_2(^1\Delta_g)$ of $2.13x10^{16}$ cm^{-3} or a yield of 16.4%. Given the simplicity of the theory, it is in reasonable agreement with our measured yields of ≈15-16% (see Section 3).

A similar calculation can be done for the $O_2(^1\Sigma_g)$ density, and it yields a smaller density by about a factor of 3x1.66 = 4.98. The factor of 3 arises because of using only ~15% of the power (as opposed to 45% for $a^1\Delta_g$) and the factor of 1.66 because it costs 1.627 eV to produce $b^1\Sigma_g$ state as opposed to 0.9775 eV for the $a^1\Delta_g$ state. The origin of the factor of 3-4 (in Figs. 4 and 5) is a consequence of the fact that the cross-section for electron impact production of is at least 4 times that for $b^1\Sigma_g$ for virtually all electron energies of significance. This ratio is in close agreement with current experimental results.

More detailed theoretical calculations of an inductive discharge utilizing Maxwell's equations (not presented for brevity) were also made. These calculations indicate that the effective E/N for typical flow conditions in our laboratory setup (10 mmol/s of pure oxygen, corresponding to a number density of $1.3x10^{17}$ cm^{-3}) should be approximately $1.2x10^{-16}$ V-cm^2 at a RF coil current of 30 amps (rms); note that this is very close to optimum, Fig. 4.

3. RECENT EXPERIMENTAL RESULTS

CUA and UIUC are currently developing the ElectriCOIL device. A two-inch flow tube experiment was constructed to demonstrate generation of $O_2(^1\Delta)$ and $O_2(^1\Sigma)$ by electrical means [King, 2001]. Key flow and electrical parameters were varied to characterize the system performance. Figure 6 depicts a block diagram of the experimental setup. RF power is transferred to the flow through a "Pi" matching network driven by a 1-kW ENI OEM-12A RF power amplifier at 13.56 MHz. Diagnostics play a critical role in developing an understanding the ElectriCOIL system; as such, a great deal of care has been taken to implement high quality diagnostics for evaluating the flow properties emerging from the discharge region.

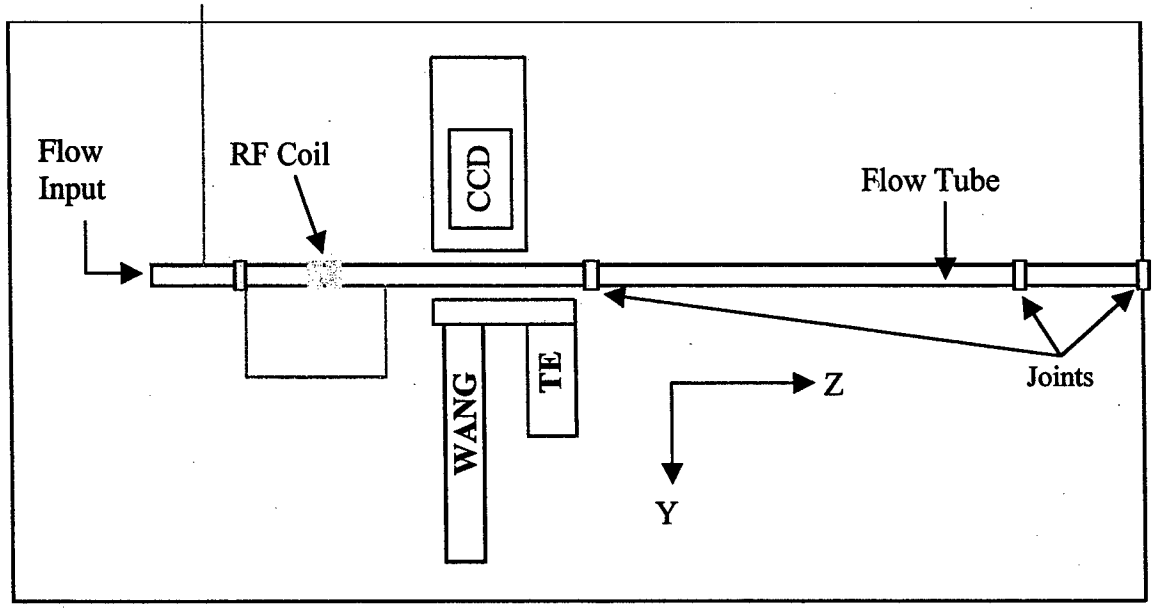

Figure 6: ElectriCOIL experimental setup at UIUC.

One of the primary measurements on the ElectriCOIL experiment is a spectrographic determination of $[O_2(^1\Sigma)]$ from its emission at 760 nm. Studies were performed to investigate the effect of oxygen and diluent flow rates, diluent type, system pressure, RF power levels, and axial flow position. The results of some of the parametric studies are presented in Fig. 7. Axial distance is measured from the end of the discharge section. Differing diluent species yielded considerably

different behaviors on the $O_2(^1\Sigma)$ levels. In Fig. 7 it is seen that the $O_2(^1\Sigma)$ level drops exponentially in the flow direction. It is of interest that the amount of Helium diluent added affects not only the concentration of the $O_2(^1\Sigma)$, but it also decreases the spatial decay downstream, an effect attributed to the increased flow velocity.

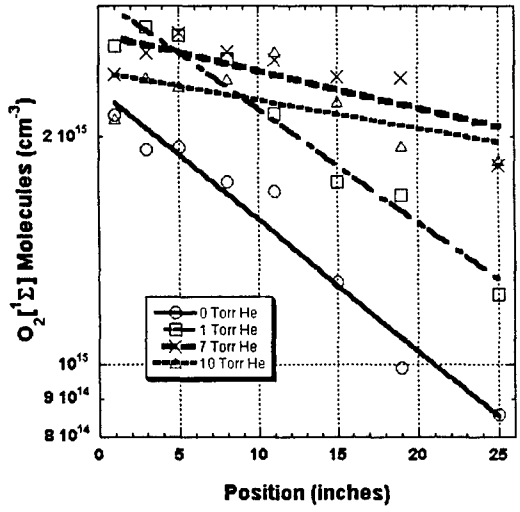

Figure 7: Effect of Helium flow rate (1.2 mmol/s of He per 1 Torr He) on $O_2[^1\Sigma]$ level with varying position for 7 mmol/s of oxygen flow. Separation of the peaks of the P and R branches of the emission indicated a gas temperature of approximately 340 K.

The next key measurement is that of the $O_2(^1\Delta) \rightarrow O_2(^3\Sigma)$ emission at 1268 nm. Results from a test setup using a sensitive Wang LN_2-cooled germanium detector and measuring the spectra with a spectrometer are shown in Fig. 8; this spectral measurement definitively verified the presence of significant percentages of $O_2(^1\Delta)$ evolving from RF discharge flows. It should be noted that the addition of NO to the discharge created a pedestal to the spectra shown in Fig. 8, but did not significantly enhance integrated $O_2(^1\Delta)$ spectra above that base. Recent measurements of the concentration of $O_2(^1\Delta)$ have been performed using a Wang detector, filtered by a CVI 1268 nm narrow bandpass filter. A great deal of care has been taken in the calibration of this measurement using two different calibrated light sources to insure the accuracy of these difficult measurements, however these are absolute intensity measurements that are inherently very difficult; the results presented in this paper are our best estimates that we believe are good to within a factor of two. Figure 9 shows that these recent measurements indicate an $O_2(^1\Delta)$ concentration of $\approx 1.9 \times 10^{16}$ cm^{-3} in a 10 mmol/s, 3.6 Torr (1.27×10^{17} cm^{-3}) flow of pure oxygen, which corresponds to a yield of ≈15%; these results are consistent with the $O_2(^1\Sigma)$ concentrations in the discharge, Fig. 7.

Based upon the laser system modeling results (Figs. 1 and 2), this measured yield of ≈15% is already at the minimum required for the ElectriCOIL system to achieve lasing. Other operating conditions have produced yields of ≈16%. Work to improve this value is continuing.

It is important to note that these yield numbers are relative to the total oxygen flow rate that is run through the discharge region. Thus, the yield as discussed so far does not reflect the fact that there is production of $O_2(^1\Sigma)$ and O atoms. Since the forward and backward reactions for the production of I* in the laser cavity region depend only upon excited $O_2(^1\Delta)$ and ground state $O_2(^3\Sigma)$ (excluding any other reactions new to the ElectriCOIL kinetics), it may be more appropriate to define an "effective yield" that is output from such discharges.

$$Y_{eff} = \left\{ \frac{[O_2(^1\Delta)]}{[O_2(^1\Delta)] + [O_2(^3\Sigma)]} \right\}_{generator\,output} . \qquad (2)$$

Measurements by Ivanov [Ivanov, 1999] suggest that the concentration of O atoms is approximately the same as that of $O_2(^1\Delta)$. Hence, for the aforementioned 3.6 Torr case (1.27×10^{17} cm^{-3}), if we account for dissociation and assume that 1×10^{16} cm^{-3} of O_2 has been converted to 2×10^{16} cm^{-3} of O atoms, assume a concentration of approximately 2.5×10^{15} cm^{-3} for $O_2(^1\Sigma)$, then a yield of ≈15% rises to an effective yield of ≈17%.

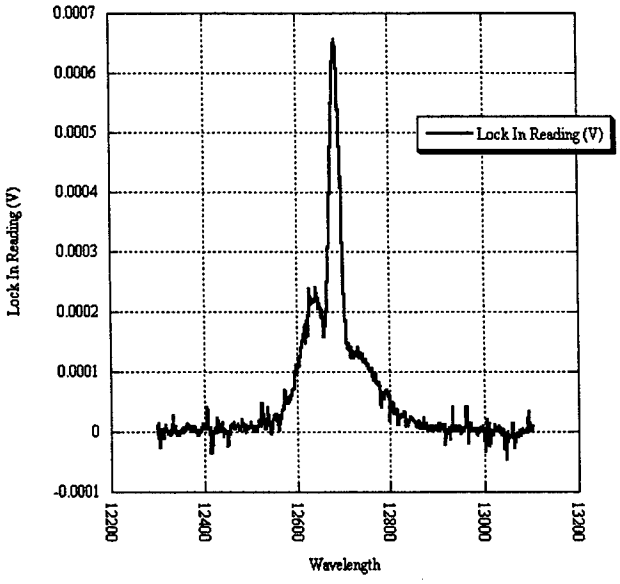

Figure 8. $O_2(^1\Delta)$ spectra taken from the ElectriCOIL RF-pumped system.

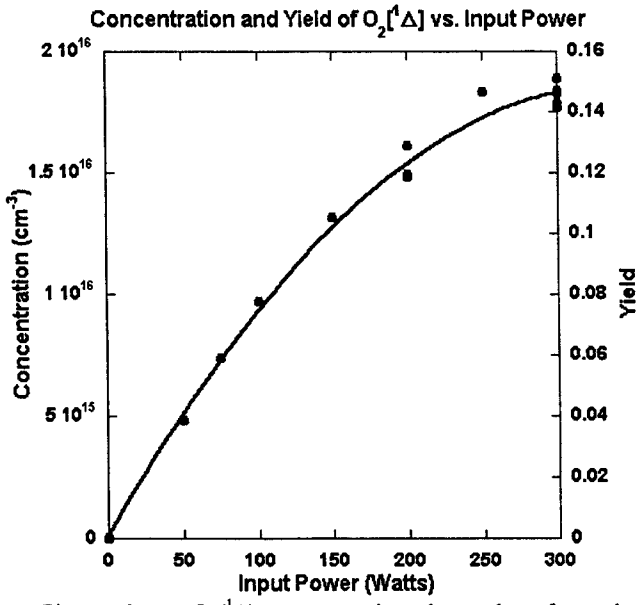

Figure 9. $O_2(^1\Delta)$ concentration data taken from the ElectriCOIL RF-pumped system at a flow rate of 10 mmol/s of pure oxygen.

Further very promising results from this research were obtained when molecular iodine was injected into the flow tube downstream of the oxygen discharge section. Two experiments were run. The flow rate of molecular iodine was approximately 0.006 mmol/s. The first had a simple 1/8" tube that was partially inserted into the flow to inject a single crossflow jet into the primary oxygen flow, Fig. 10. The second test utilized a 1/8" tube with 6 small holes drilled in the side; molecular iodine was injected in a direction parallel to the primary oxygen flow, Fig. 11. In both cases, the yellow glow from the $I_2(B) \rightarrow I_2(x)$ transition was immediately observed. The yellow glow is definitive evidence of the presence of significant concentrations of $O_2[^1\Delta]$.

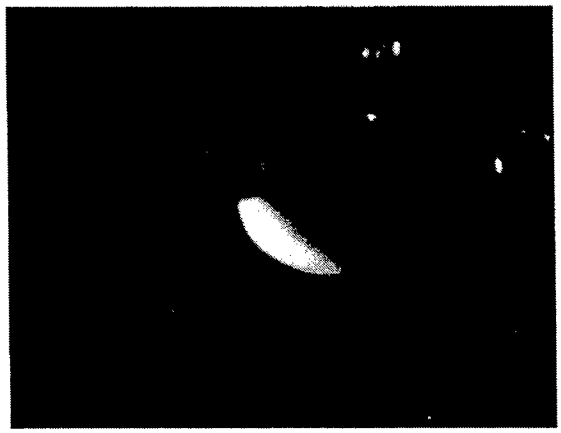

Figure 10. Single jet iodine injection.

Figure 11. Multi-jet iodine injection.

4. SUMMARY

Advanced chemical iodine laser technology will logically include novel all gas phase generation techniques for an iodine energy donor and the injection of atomic rather than molecular iodine. A candidate method, RF excitation, has been

investigated in this paper. It is seen that both the singlet-sigma $O_2(^1\Sigma)$ and singlet-delta $O_2(^1\Delta)$ excited states of the oxygen molecule may be readily created via electrical excitation in the laboratory. Generally an RF discharge alone is sufficient to bring about this excitation. However, at higher flow rates (>15 mmol/s) it has been found that both an ac discharge used in tandem with the RF discharge is important to achieve high levels of excitation [King, 2001]. We believe that it may be possible to successfully supplant the two-phase elements (liquid BHP and a chlorine/helium/nitrogen gas mixture) of the classical COIL SOG using the ElectriCOIL prototype system.

Diagnostics play a critical role in developing an understanding the ElectriCOIL system; as such a great deal of care has been taken to implement high quality diagnostics for evaluating the flow properties emerging from the discharge region. Recent measurements indicate an $O_2(^1\Delta)$ concentration of approximately $2x10^{16}$ cm^{-3} in a 10 mmol/s, 3.6 Torr ($1.3x10^{17}$ cm^{-3}) flow of pure oxygen, which corresponds to a yield of ≈15%; these results are consistent with measured $O_2(^1\Sigma)$ levels of $2.5x10^{15}$ cm^{-3} in the discharge and theoretical estimates. Other operating conditions have produced yields of ≈16%. We are planning to conduct experiments with additional advanced diagnostics in the near future.

ElectriCOIL will reduce weight and simplify both military and commercial chemical iodine laser systems. Potential cost and weight savings are also envisioned as the massive quantities of liquid chemicals will be completely eliminated from the device operation. Difficulty will certainly be encountered when searching for a yardstick to predict ElectriCOIL performance. Typical measures such as chemical efficiency must be redefined (or eliminated) to account for changes in the chemistry and parameters like the electrical power absorbed by the flow from the RF discharge. Sets of diagnostics and analysis are planned to take advantage of the recent progress in our laboratories.

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